Impact modified polybutylene terephthalate

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Inventor(s): VAN DER MEER ROELOF (NL); GOFF LESLIE JOE (NL); VAN OOSTERBOSCH CORNELIS JACOB (NL)

Applicant(s):: GEN ELECTRIC (US)

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Abstract

The invention provides a polybutylene terephthalate composition which contains an impact modifier in an impact modifying amount, wherein the impact modifier is a synergistic mixture of (1) an acrylonitrile and butadiene unit-containing polymer and (2) a terpolymer of (a) ethylene, (b) a lower alkylacrylate and (c) a monomer which contains a heterocycle containing one oxygen atom as the hetero-atom.

Description

This invention relates to impact modified polybutylene terephthalate.

Poly(butylene terephthalate) (hereinafter abbreviated as PBT) is a thermoplastic material having good chemical resistance and relatively good flow. PBT possesses a glass transition temperature of approximately 40-50 DEG C. Therefore, in order to achieve the toughness needed for most practical purposes, an impact modifier should be added to the PBT. Several impact modifiers have been used for PBT previously, including ABS which is customarily used in PBT compositions not containing polycarbonate. A special kind of ABS-graft polymers has been disclosed as impact modifiers for PBT in EP-A-0022216. Also acrylonitrile-butadiene copolymers have been used for this purpose and according to Fowler, M.E. Keskkula, H, Paul, D.R., Polymer, 28, p.1703-1711 (1987) copolymers from acrylonitrile and cis-butadiene provide a low temperature ductility. A further problem is that for some uses good charpy notched impact strength (hereinafter sometimes abbreviated as CNI) is desired. For CNI there is a very strong concentration effect caused by the notch in this test, and therefore the material strain is constrained entirely in the middle of the specimen bar, and around the fracture surface, as discussed by Polato, F., J.Mat.Sci. 20, p. 1455-1465, (1985). A CNI of at least 25 J/m which is needed for some purposes, can be obtained with high percentages of ABS polymers or the like, but in that case the viscosities suffer severely. Accordingly, there is a need for an impact modifier for PBT which in moderate contents provides the desired high CNI values.

Surprisingly it has now been found that a group of specific terpolymers has somewhat better impact modifying properties than the acrylonitrile-butadiene copolymers and terpolymers, and more particularly that combinations of these specific terpolymers with the acrylonitrile-butadiene copolymers and terpolymers show a completely unexpected synergistic action so that with these combinations CNI values at room temperature of 25 J/m2 and above can easily be attained.

Thus, the invention provides an impact modified polybutylene terephatalate composition containing an impact modifier in an impact modifying amount, wherein the impact modifier is a synergistic mixture of (1) an acrylonitrile and butadiene units-containing polymer and (2) a terpolymer of (a) ethylene, (b) a lower alkyl acrylate and (c) a monomer which contains a heterocycle containing one oxygen atom as the hetero-atom.

Component (1) is one of the usual impact modifiers for this purpose and can be an acrylonitrile-butadiene copolymer or one of the several ABS polymers, for instance of the graft or core-shell type.

Component (2) is a terpolymer wherein the greater parts of the monomers (55-75 %) is derived from ethylene, 20-30 % of the monomers is derived from a lower alkyl acrylate, and 5-15 % of the monomers is derived from monomer (c), it being understood that the sum of the three percentages is 100. By "lower alkyl acrylate" is understood here a C1-4 alkyl ester of acrylic or methacrylic acid.

Component (c) can contain the heterocycle as part of the main polymer chain or in a pendant group. An example of the first possibility is maleic anhydride, and a particularly suitable pendant heterocylic group is the glycidyl residue. A particularly preferred monomer (c) is glycidyl methacrylate.

Some of these terpolymers are commercially available and all these terpolymers will be produced according to well-known processes.

The weight ratio between components (1) and (2) can vary within wide limits, but is preferably 10:1 to 1:1.5 and more preferably 4:1 to 1:1.

As mentioned hereinabove, the impact modifying synergistic mixture should be used in an impact modifying amount. This amount can also vary within wide limits, and will also depend on the influence on other physical properties which can be tolerated. In general the synergistic combination of the present invention will be used in a total amount of 6-20 weight percent of the composition. Amounts of less than 6 % usually will not have sufficient impact modifying effect and amounts of substantially over 20 % will tend to unduly affect other desirable properties. Preferably the amount of the synergistic combination is 10-15 % of the entire composition.

Of course, the compositions of the invention may contain minor amounts of further usual additives, such as anti-oxidants, fillers, reinforcing fillers, stabilizers, dyes and pigments, plasticizers, mold release agents, lubricants, antistatic agents, flame retardants and the like.

The following examples are intended to elucidate the present invention. However, the invention is not limited by these examples in any way.

Example 1

PBT compositons were prepared with various impact modifiers, and the physical properties of the compositions were measured. The results are as follows:

These screening tests show that when using the ABS polymer as impact modifier, CNI values of over 25 kJ/m2 can only be obained with high percentages of the ABS with concomitant unacceptably low MVI values. 10 % of terpolymer (A) gives a clearly higher CNI value than the same amount of the ABS, and terpolymer B in a content of 10 % gives a considerably higher CNI value. The two other tested substances give worse results than the ABS, when used in the same amount of 10 %.

Example 2

Combinations of the ABS and the terpolymer B of Example 1 were tested in three different ratios. The results are given in the following Table.

As appears from these results the desired CNI values of over 25 kJ/m2 are easily attained with the three tested mixtures. It should be remarked in this respect that terpolymer B is a more expensive material than the ABS so that it is economically very interesting that approximately the same CNI values were obtained over the examined range of ABS/terpolymer B ratios of 1:1 till 4:1. The commercial product used as a comparison is believed to contain also 15 % of impact modifier.

The reason for the unexpected synergism of the present invention is not known with certainty.

All patent (applications) mentioned herein are herewith incorporated by reference.

Claims

1. Polybutylene terephthalate composition containing an impact modifier in an impact modifying amount, wherein the impact modifier is a synergistic mixture of (1) an acrylonitrile and butadiene units-containing polymer and (2) a terpolymer of (a) ethylene, (b) a lower alkylacrylate and (c) a monomer which contains a heterocycle containing one oxygen atom as the hetero-atom.

2. Polybutylene terephthalate composition according to claim 1, wherein component (1) is a copolymer of acrylonitrile and butadiene.

3. Polybutylene terephthalate composition according to claim 1, wherein component (1) is an ABS polymer.

4. Polybutylene terephthalate composition according to claims 1-3, wherein monomer (c) is glycidyl methacrylate.

5. Polybutylene terephthalate composition according to claims 1-3, wherein monomer (c) is maleic anhydride.

6. Polybutylene terephthalate composition according to claims 1-5, wherein the weight ratio of component (1) and component (2) is 10:1 to 1:1.5.

7. Polybutylene terephthalate composition according to claim 6, wherein the ratio is 4:1 tot 1:1.

8. Polybutylene terephtalate composition according to claim 1 comprising further usual additives.

9. Polybutylene terephthalate composition according to any claims 1-8, wherein the synergistic mixture comprises 6-20 weight percent of the entire composition.

10. Polybutylene terephthalate composition according to claim 9, wherein the synergistic mixture comprises 10-15 weight percent of the entire composition.

11. Polybutylene terephthalate composition according to any of claims 1-10 having a charpy notched impact strength at room temperature of at least 25 J/m2.
